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1                   **Characterization of sulfate mineral deposits in central Thailand**

2

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21

22 **Abstract**

23 In this paper we present petrographic and geochemical data of sulfate mineral  
24 deposits in northeast Nakhon Sawan, central Thailand, and provide new constrains on  
25 their age. The deposits are made up mainly of strongly deformed nodular and massive  
26 gypsum in the upper part, and less deformed layered anhydrite in the lower part. They  
27 have been intruded by andesitic dikes that contain Middle Triassic zircons (~240 Ma).  
28 These dikes are probably part of the regional magmatic activity of the Sukhothai Arc  
29 during the Early to Middle Triassic. Sulfur ( $\delta^{34}\text{S}$ ) and strontium ( $^{87}\text{Sr}/^{86}\text{Sr}$ ) isotopic  
30 compositions of the sulfates range from 15.86‰ to 16.26‰ and from 0.70810 to  
31 0.70817, respectively. Comparisons with the Phanerozoic seawater isotopic evolution  
32 curve indicate that those values are best explained by precipitation of the sulfates from  
33 Carboniferous seawater, in particular seawater of late Mississippian age (~326 Ma), and  
34 this would be consistent with previous studies of calcareous fossils in the limestones  
35 that crop out around this site. Our interpretation is that evaporitic gypsum was originally  
36 precipitated from hypersaline seawater on a shallow lagoon or shelf on the Khao  
37 Khwang Platform during the Serpukhovian, and that this gypsum changed to anhydrite  
38 during early burial. The anhydrite was then cut by andesitic dikes during the Middle  
39 Triassic, and more recently the upper part of which was rehydrated during exhumation  
40 to form secondary gypsum near the surface.

41

42 **Key words:** sulfate evaporites, strontium isotopes, sulfur isotopes, Carboniferous

43

44 **INTRODUCTION**

45 Calcium sulfate deposits (gypsum and anhydrite) of Paleozoic age are distributed  
46 around the Loei–Wang Saphung and Nakhon Sawan areas of mainland Thailand (e.g.,  
47 Jacobson *et al.* 1969; Chonglakmani *et al.* 1983; El Tabakh & Utha–Aroon 1998; Ueno  
48 & Charoentitirat 2011). Among these, the sulfate deposits in the Loei–Wang Saphung  
49 area of northeast Thailand have been investigated in drilled cores (Utha–Aroon &  
50 Surinkum 1995; Surakotra *et al.* 2005; Surakotra 2011). Based on the occurrence of  
51 fusulinids from a limestone interval above the gypsum–anhydrite deposits, a late  
52 Moscovian (Pennsylvanian/late Carboniferous) age has been proposed for the sulfate  
53 deposits (Fontaine *et al.* 1997). However, the age of the gypsum–anhydrite itself still  
54 remains unknown, because the stratigraphic relationship between the sulfate deposits  
55 and the fossil-bearing limestone is unclear from borehole data (Surakotra *et al.* 2005).  
56 Large exposures of the sulfate deposits are found in quarries in northeastern Nakhon  
57 Sawan Province of central Thailand (Fig. 1), and some are still exploited by mining  
58 companies. In contrast to the Loei–Wang Saphung area, only a few studies have  
59 examined the deposits in the Nakhon Sawan area, and as yet there is no direct evidence  
60 for their age.

61 In this paper we characterize the sulfate mineral deposits based on petrographic  
62 observations, major and trace element compositions, and isotopic compositions of the  
63 sulfur ( $\delta^{34}\text{S}$ ) and strontium ( $^{87}\text{Sr}/^{86}\text{Sr}$ ) in order to assess their origin, the processes of  
64 precipitation, and the nature of diagenetic alteration. If the deposits were precipitated  
65 from seawater, their  $\delta^{34}\text{S}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  values could be used to constrain their age by  
66 making comparisons with the global reference curves of  $\delta^{34}\text{S}$  (Kampschulte & Strauss  
67 2004) and  $^{87}\text{Sr}/^{86}\text{Sr}$  (McArthur *et al.* 2012) for Phanerozoic seawater. Sr isotopic record

68 is particularly useful because of well-homogenized isotopic composition through the  
69 global oceans due to its long residence time. Although  $\delta^{34}\text{S}$  values are different for  
70 sulfate evaporites and carbonate-associated sulfur, they basically show parallel trends  
71 with offsets that are generally less than 4‰ through the Phanerozoic (Kampschulte &  
72 Strauss 2004). The seawater  $^{87}\text{Sr}/^{86}\text{Sr}$  curve has been compiled for the Phanerozoic by  
73 McArthur *et al.* (2012) and is stored in a database called LOWESS.

74 The sulfate rock bodies are intruded by igneous dikes. We investigated petrography  
75 and geochemistry of the igneous dikes, and conducted U-Pb dating of zircons separated  
76 from the igneous intrusions to constrain the upper (younger) limit of the formation age  
77 of the sulfate rock body.

78

## 79 **GEOLOGICAL SETTING**

80 The basement rocks in Thailand can be divided into three geotectonic units; from  
81 west to east, the Sibumasu Block, the Sukhothai Zone, and the Indochina Block (Ueno  
82 & Charoentitirat 2011) (Fig. 2 A). The Sibumasu Block represents the peri-Gondwanan  
83 terrane, containing upper Carboniferous to lower Permian glaciogenic diamictites with  
84 Gondwanan fauna and flora, and middle–upper Permian platform carbonates. On its  
85 eastern side, a peculiar geotectonic domain called the Inthanon Zone can be defined; in  
86 this zone, Paleo-Tethyan oceanic rocks, made up of Carboniferous to Permian seamount  
87 carbonate rocks associated with greenstones and Middle Devonian to Middle Triassic  
88 radiolarian cherts, are widely exposed as tectonic outliers that rest on the underlying  
89 pre-Devonian basement (Cambrian sandstone and Ordovician limestone). The  
90 Sukhothai Zone is mainly made up of deformed Paleozoic to Mesozoic sedimentary and  
91 volcanic rocks. Triassic I-type granitoids are also present, and they suggest a

92 subduction-related tectonic setting and a volcanic arc that developed along the margin  
93 of the Indochina Block. To the east, upper Paleozoic shallow-marine carbonate rocks  
94 with diverse Tethyan faunas are widely distributed along the western margin of the  
95 Indochina Block. The Indochina Block, which occupies the eastern half of mainland  
96 Thailand, is part of the South China and Indochina Superterrane (Metcalf 2000). It has  
97 been considered to be within the paleo-equatorial region since its Early Devonian rifting  
98 from Gondwana.

99       In the eastern part of the Nakhon Sawan area, the sedimentary rocks consist mainly  
100 of 1) Permian carbonate rocks of the Tak Fa Formation of the Saraburi Group, and 2)  
101 small amounts of Carboniferous strata (Fig. 2 B). These sedimentary rocks are widely  
102 distributed along the western margin of the Khorat Plateau of northeast Thailand, and  
103 this plateau is a part of the Indochina Block (El Tabakh & Utha–Aroon 1998; Ueno &  
104 Charoentitirat 2011; Ueno *et al.* 2012). The Carboniferous rocks have been found in  
105 several locations around Chon Daen, east of Nakhon Sawan (Chonglakmani *et al.* 1983;  
106 Fontaine *et al.* 1983; Ueno & Charoentitirat 2011), and they can be described as follows.  
107 A basal chert–volcanic tuff succession is overlain by dark-gray limestone that locally  
108 contains abundant foraminifera and corals of Visean age (Fontaine *et al.* 1991). In turn,  
109 the limestones are overlain by shales, siltstones, sandstones, and thin limestone beds and  
110 lenses. The shales and siltstones contain trilobites, brachiopods, and solitary corals, and  
111 the thin limestone beds yield Visean foraminifera. In this area several large sulfate  
112 deposits (approximately 10 km × 10 km areal extent) are exposed in active quarries (Fig.  
113 1), and the deposits are crosscut by andesitic dikes. The distribution of sulfate mineral  
114 deposits is closely associated with reddish shales and sandy shales that appear to overlie  
115 the Visean strata (Chonglakmani *et al.* 1983). This implies that the timing of sulfate

116 precipitation was close to the Mississippian–Pennsylvanian boundary (Ueno &  
117 Charoentitirat 2011), although direct age constraints have not yet been obtained.

118

## 119 MATERIALS AND METHODS

120 Investigated sulfate rock samples were taken from three quarries in the Chon Daen  
121 area, east of Nakhon Sawan (Fig. 1). In the Chao Phraya Mueang Rae quarry in the  
122 central part of this area (Fig. 1 D), sample 13112601 was taken from below an igneous  
123 dike and sample 13112602 from the contact between the dike and the sulfate deposit;  
124 samples 13112603 and 13112603' were taken from above the dike (Fig. 3 A). In the  
125 K-Mining quarry in the northern part of this area (Fig. 1 C), sample 13112604 was part  
126 of a sulfate mineral deposit, and sample 13112605 was from an andesitic dike, collected  
127 for zircon U–Pb dating (Fig. 3 B). Each sample was cut and polished before making thin  
128 sections using the dry method proposed by Owada *et al.* (2013) for fragile samples. In  
129 the SCG Mine in the southern part of this area (Fig. 1 E), more than 20 cores have been  
130 drilled previously, and some of them are stored in this company. Sediment in a core  
131 GYP13 comprises mainly of reddish to light gray soil and clay as overburden (0–12.7 m  
132 depth), heavily deformed, white to gray gypsum (12.7–37.4 m depth), and layered gray  
133 anhydrite (37.4–60.0 m depth) (Fig. 4 A). All other cores show similar sedimentary  
134 sequence, although none of them has reached the bottom of the anhydrite. We took  
135 whole round core samples (10–20 cm long) from the GYP13 core (Fig. 4 B). In some  
136 other wells (GYP5 and GYP9), andesitic dikes cut both gypsum and anhydrite. After  
137 removing outer part of each sample block or core to avoid any contamination, all  
138 samples were pulverized using an agate mortar in preparation for bulk-rock analysis.

139 The distributions of major elements (Na, Mg, Al, Si, S, K, Ca, Fe, and Sr) in an area

140 of 20 mm × 30 mm in thin sections was determined by X-ray elemental mapping using  
141 a JEOL JXA-8500F field-emission electron probe microanalyzer (FE-EPMA) at Japan  
142 Agency for Marine-Earth Science and Technology (JAMSTEC). Measurements were  
143 performed with a 15 kV accelerating voltage, a 50 nA specimen current, and a focused  
144 beam of <1 μm diameter. Characteristic X-rays of all elements were measured from  
145 K-lines using a wavelength-dispersive spectrometer (WDS). Elemental compositional  
146 maps were obtained by scanning samples in stages, counting X-rays at each pixel, and  
147 with a 40 ms counting time. Details of the methods of sample preparation and the  
148 measurement conditions are given in Kuroda *et al.* (2005).

149 For bulk-rock major element, trace element, and Sr isotope analyses, samples were  
150 dissolved overnight with ~30% HCl at 120 °C. After heating to dryness, each sample  
151 was re-dissolved in 3% HNO<sub>3</sub>. Subsamples were taken for inductively coupled plasma-  
152 mass spectrometry (ICP-MS) analysis, and diluted further with 3% HNO<sub>3</sub>. Major and  
153 trace element concentrations were measured using a Thermo iCap quadrupole ICP-MS.  
154 Calibrations for each element have been given by Kuroda *et al.* (2007, 2016) and Hara  
155 *et al.* (2010). Analytical uncertainties (as relative standard deviation) for the bulk rock  
156 analysis were better than 3% for major elements, and better than 5% for trace elements.

157 Concentrations and stable isotopic compositions of sulfur were determined in  
158 powdered samples using an EA/IRMS (elemental analyzer/isotope-ratio mass  
159 spectrometer) system (Isoprime-EA, Isoprime) at the University of Tsukuba. The  
160 analytical procedures and conditions have been described by Maruoka *et al.* (2003). The  
161 sulfur isotopic compositions are expressed in terms of δ<sup>34</sup>S (‰) relative to the Vienna  
162 Canyon Diablo Troilite (V-CDT) standard. The isotopic compositions of sulfur were  
163 determined with a precision of ± 0.2‰ (1σ). Two standard reference materials for sulfur

164 (IAEA-S-1 and 2) were used for calibrating sulfur contents and for correcting the  
165 instrumental mass discrimination of the IRMS.

166 Separation of Sr for isotopic analysis was carried out at JAMSTEC, using a method  
167 described by Roveri *et al.* (2014), with slight modifications. Sr was separated using  
168 Eichrom Sr Spec resin. Matrix elements were eluted in 6M HNO<sub>3</sub> and 3M HNO<sub>3</sub> before  
169 collecting Sr with 0.05M HNO<sub>3</sub>. The total procedural blank for Sr samples prepared  
170 using this method was less than 10 pg. Samples were loaded onto single Re filaments  
171 with a Ta-activator (Takahashi *et al.* 2009), and the Sr isotopic composition of each  
172 sample was measured with a Thermo Scientific TRITON TI thermal ionizing mass  
173 spectrometer at JAMSTEC (Takahashi *et al.* 2009). The data were acquired in static  
174 multi-collection mode, and computed from 10 blocks of 15 cycles with an integration  
175 time of 15 sec for each cycle. The <sup>87</sup>Sr/<sup>86</sup>Sr ratio was normalized for mass fractionation  
176 using an exponential law correction to the <sup>86</sup>Sr/<sup>88</sup>Sr ratio of 0.1194. Analytical accuracy  
177 was evaluated by measuring NIST SRM 987, which provided readings of 0.7102455 ±  
178 0.0000011 (2 S.D., *n* = 3) during the course of this study. The 2 standard error value of  
179 internal precision on an individual analysis was between 0.000008 and 0.000012. We  
180 did not correct for interference of <sup>87</sup>Rb because <sup>87</sup>Rb was not detected. The Sr isotope  
181 measurements were performed with an <sup>88</sup>Sr ion beam intensity of 3–5 V.

182 U–Pb dating was carried out for zircons in the andesitic dike sample 13112605 (Fig.  
183 1 C). Zircons were separated using isodynamic magnetic and sodium polytungstate  
184 (SPT) heavy liquid separation techniques. Zircon grains were picked randomly from  
185 this concentrate, and mounted on and embedded in a Teflon sheet.  
186 Cathodoluminescence (CL) imaging was performed to observe the internal structures  
187 and zonation patterns of the zircons, and to select suitable sites for U–Pb dating using

188 SEM–EDS (JEOL JSM-6610 LV) equipped with a CL system (Gatan Mini CL) at the  
189 Geological Survey of Japan (GSJ). For the U–Pb age determinations, the U–Pb isotope  
190 abundance ratio was analyzed using laser ablation quadrupole–type ICP–MS  
191 (Agilent7700x, housed at Nagoya University), following the methods of Orihashi *et al.*  
192 (2008) and Kouchi *et al.* (2015). Analyses were performed with a spot size of 25  $\mu\text{m}$ , a  
193 10 Hz repetition rate, an energy density of 11.7 J  $\text{cm}^{-2}$ , and using a 213 nm solid state  
194 laser ablation system (ESi/New Wave Research NWR213). Calibration and data quality  
195 control were undertaken using standard zircons of 91500 and NIST SRM 610 at GSJ.  
196 All ages were calculated and concordia diagrams were made using Isoplot v.3.75  
197 (Ludwig 2012).

198

## 199 **RESULTS**

### 200 **PETROGRAPHIC CHARACTERISTICS OF THE SULFATE DEPOSITS**

201 The sulfate rocks consist mostly of heavily sheared gray to light gray massive  
202 microcrystalline gypsum (Fig. 3). **In the drilled cores at the SCG Mine, the gypsum is**  
203 **underlain by layered gray anhydrite (Fig. 4 A).** Unlike to the overlying gypsum, the  
204 anhydrite beds are much less deformed (Fig. 4 B). This implies that the top part of the  
205 anhydrite body has been rehydrated to gypsum due to exhumation, and that the heavily  
206 sheared structures in the gypsum samples were formed by recrystallization of anhydrite  
207 (i.e., volume increase due to rehydration). Same process has been reported from another  
208 sulfate rock body in Loei-Wang Saphung area (Surakotra, 2011).

209 In some cases a faint layering can be recognized in the gypsum and anhydrite, made  
210 evident by thin microcrystalline carbonate layers. Veneers of this carbonate outline  
211 vertically aligned centimetric gypsum nodules that resemble pseudomorphs of vertically

212 aligned selenite crystals. However, the nodules appear to have been strongly sheared  
213 during deformation and flow of the sulfate rock, and the original sedimentary features  
214 seem to have been completely obliterated.

215 Sample 13112601 consists mainly of microcrystalline gypsum with minor amounts  
216 of granular xenotopic gypsum and prismatic idiotopic gypsum. Very thin streaked-out  
217 fragments of silicate minerals have been deformed to form S-shape fold patterns (Fig. 5  
218 A). Gypsum grains have been deformed by ductile flow, while laminae of silicate  
219 minerals have been partly disaggregated by brittle deformation into granules. The  
220 textures indicate that the gypsum originated by the hydration of former anhydrite (Lugli  
221 2001).

222 Sample 13112602, which was taken from the contact zone between an andesitic  
223 dike and the sulfate deposit, is composed of xenotopic granular to idiotopic prismatic  
224 gypsum crystals up to 3 mm in size (Fig. 5 B). Deformed patches of microcrystalline  
225 gypsum a few mm in size are also present. Streaked-out fragments of silicate minerals  
226 are crosscut by veins of fibrous and prismatic gypsum crystals. As with sample  
227 13112601, the gypsum crystals in 13112602 have been deformed by ductile flow, while  
228 the silicate layers have been boudinaged due to brittle deformation.

229 Sample 13112603 is laminated and consists of xenotopic granular to idiotopic  
230 prismatic gypsum crystals (Fig. 5 C) up to 3 mm in size. Some beds have vertically  
231 aligned crystals, especially the larger crystals. Other beds have a random orientation of  
232 crystals. Smaller crystals may be aligned to form thin, vertical or oblique linear  
233 structures up to 3 mm long. Again, this gypsum rock has textures that originated by the  
234 hydration of precursor anhydrite.

235

## 236 GEOCHEMICAL CHARACTERISTICS OF THE SULFATE DEPOSITS

237 Concentrations of calcium (Table 1) and sulfur (Table 2) in gypsum samples range  
238 from 19.5 to 25.0 wt% and 16.3 to 17.1 wt%, respectively, which are typical of gypsum  
239 (~23 wt% Ca and ~19 wt% S). In contrast, the anhydrite samples have slightly higher  
240 Ca concentrations, ranging from 25.4 to 29.5 wt% (Table 1), which are close to the  
241 typical Ca value of anhydrite (~29 wt% Ca).

242 As comparison, we also measured the major and trace elements of gypsum from the  
243 Saline di Trapani, Sicily (JTG), where the gypsum has been artificially precipitated  
244 from present-day Mediterranean seawater (Table 1). The gypsum and anhydrite samples  
245 from Nakhon Sawan have lower concentrations of Mg and Sr than the gypsum from the  
246 Saline di Trapani, and these cations might have been leached out through dehydration  
247 from pristine gypsum to anhydrite during burial, and through rehydration of anhydrite to  
248 gypsum in a near-surface environment (e.g., the sulfate cycle of Murray 1964).

249 Primary anhydrite can only precipitate under relatively high temperatures and high  
250 salinity conditions as in supratidal sabkha evaporites (Warren 1999). However, our  
251 anhydrite samples show thin beddings (Fig. 4 B) suggesting a subaqueous origin rather  
252 than a supratidal evaporite. Therefore, the anhydrite beds seem to have formed  
253 secondary. The overlying gypsum is strongly deformed and recrystallized, and does not  
254 show original evaporite sedimentary structures such as selenite or twinned gypsum  
255 crystals, as commonly observed in pristine deposits (e.g., Lugli *et al.* 2010). Based on  
256 the lines of evidence we propose that the anhydrite was formed during burial at around  
257 52 °C due to dehydration of the pristine evaporitic gypsum (e.g., Murray 1964;  
258 Shearman 1980; Testa & Lugli 2000). The lithological and mineralogical features of the  
259 Nakhon Sawan sulfate deposits resemble those of the Loei–Wang Saphung area, as

260 described by Surakotra *et al.* (2005), who suggested that the primary sulfate mineral  
261 was bedded selenite gypsum, precipitated on the floors of lagoons, lakes, or shallow  
262 shelves under arid climatic conditions. Likewise, we interpret that the sulfate deposits of  
263 Nakhon Sawan were originally precipitated in a shallow marine environment on the  
264 floor of a lagoon or shelf.

265

#### 266 GEOCHEMICAL CHARACTERISTICS OF THE IGNEOUS DIKES

267 The dikes are light to dark green in color, and 1–1.5 m in width, and they were  
268 intruded into the gypsum along vertical fractures that strike 20 °E at the sampling site of  
269 13112605 in the northern quarry (Fig. 3 B). The SiO<sub>2</sub> and Na<sub>2</sub> + K<sub>2</sub>O contents of one of  
270 these dikes are 59.91 wt% and 2.03 wt%, respectively (Supporting Information Table  
271 S1), and the rock plots in the andesite field on the discrimination diagram of Wilson  
272 (1989). The andesitic dike is mainly made up of saussuritized plagioclase and small  
273 fractured phenocrysts of pyroxene (Fig. 5 D), and is locally cut by quartz and calcite  
274 veins. The groundmass contains glassy black seams, and is commonly altered to chlorite.  
275 The dike also contains xenoliths of dacitic, andesitic, and basaltic volcanic rocks, as  
276 well as abundant xenocrysts of quartz (Fig. 5 D).

277 At the sampling site of 13112602 in the southern quarry, an andesitic dike that  
278 intruded the gypsum deposit is composed mainly of plagioclase phenocrysts, is several  
279 to tens of centimeters in width, and has a horizontal attitude (Fig. 3 A). In sample  
280 13112602, the dike exhibits laminae-like structures in the element maps of Mg, Si, and  
281 Fe (Fig. 6). The dike is characterized by cracks that are filled with gypsum, and this  
282 cracking might have occurred by volume expansion due to the rehydration of anhydrite  
283 to gypsum, with pore spaces filled with secondary gypsum.

284

## 285 SULFUR ISOTOPE RATIOS OF THE GYPSUM

286 The sulfur isotopic compositions ( $\delta^{34}\text{S}$ ) of the gypsum fall in a narrow range  
287 between 15.86‰ and 16.26‰ (Table 2). Worden *et al.* (1997) demonstrated that the  
288 dehydration of gypsum into anhydrite does not involve significant isotopic fractionation  
289 or diagenetic redistribution of material. They showed that the stratigraphic variations of  
290 primary seawater  $\delta^{34}\text{S}$  values in the Permian–Triassic were preserved in anhydrites in  
291 Abu Dhabi, despite post-depositional dehydration occurring at burial depths greater than  
292 ~1000 m. Although isotopic fractionation may have occurred on the submillimeter scale  
293 at Abu Dhabi, it was averaged out on the scale of individual centimeter-scale anhydrite  
294 nodules. We interpret that this was also the case for the gypsum samples at Nakhon  
295 Sawan. Sulfur isotopic compositions may also have been modified by the intrusion of  
296 the igneous dike. However, the  $\delta^{34}\text{S}$  values of the sample nearest to the igneous dike  
297 (13112602) are identical to those located >20 m from the dikes (e.g., 13112604) (Table  
298 2). This suggests that isotopic fractionation associated with the magmatic activity was  
299 negligible.

300 Because sulfur isotopic fractionation at the time of precipitation of sulfates from  
301 seawater is generally very small (e.g., Raab & Spiro 1991), we interpret the  $\delta^{34}\text{S}$  values  
302 as primarily reflecting those of the co-existing seawater (brine), in other words, the  $\delta^{34}\text{S}$   
303 values of the gypsum can be correlated with the  $\delta^{34}\text{S}$  values of contemporaneous ocean  
304 water sulfates (e.g., Paytan & Gray 2012).

305

## 306 STRONTIUM ISOTOPE RATIOS OF THE GYPSUM

307 Like sulfur isotopic compositions, the  $^{87}\text{Sr}/^{86}\text{Sr}$  values of the gypsum and anhydrite

308 samples fall in a narrow range between 0.70811 and 0.70817 (Table 3). This strongly  
309 suggests that Sr isotopic composition is not influenced through the rehydration process  
310 from anhydrite to gypsum. In addition, it is also evident that andesitic intrusion did not  
311 alter the Sr isotopic composition of sulfate minerals significantly, because the  $^{87}\text{Sr}/^{86}\text{Sr}$   
312 value of the sample 13112603 which was taken nearby the intrusion also falls in the  
313 narrow range (0.70811). Sr isotopic compositions of evaporitic sulfates are known to be  
314 identical to those of ocean water, provided the brine (saline seawater) is fully connected  
315 with the ocean water. For example, the  $^{87}\text{Sr}/^{86}\text{Sr}$  values of the primary lower gypsum  
316 that formed in the Mediterranean Sea during the first stage of the Messinian Salinity  
317 Crisis (5.97 to 5.61 Ma; Roveri *et al.* 2014) are almost identical to those of the coeval  
318 ocean water ( $\sim 0.7090$ ; McKenzie *et al.* 1988; McArthur *et al.* 2012), but show a slight  
319 decreasing trend (e.g., Müller & Mueller 1991; Flecker & Ellam 1999, 2006; Flecker *et*  
320 *al.* 2002; Roveri *et al.* 2014). This suggests that the dissolved Sr inventory in the  
321 Mediterranean was dominated by ocean water, but that there were minor but progressive  
322 contributions of Sr, fed into the sea from adjacent drainage basins. Much lower  $^{87}\text{Sr}/^{86}\text{Sr}$   
323 values have been observed for gypsum samples formed during the second and third  
324 stages of the salinity crisis (5.61 to 5.33 Ma; Roveri *et al.* 2014), when the connection  
325 of the Mediterranean with the global ocean was significantly restricted. In the second  
326 stage of the salinity crisis, thick halite was deposited in the deep basins, while the third  
327 stage is characterized by more significant influences of brackish water in the  
328 Mediterranean basins (e.g., CIESM 2007). Under the restricted basin conditions, Sr was  
329 mostly supplied from rivers, and the contribution of ocean water was diminished during  
330 these stages.

331 The sulfate mineral deposits in the Nakhon Sawan area are dominated by gypsum

332 and anhydrite, whereas halite, polyhalite, and kainite, which only precipitate from much  
333 denser saline water, are absent. Therefore, we interpret that the sulfate mineral deposits  
334 were precipitated from seawater that was still connected with ocean water, similarly to  
335 the primary lower gypsum deposits of the Mediterranean marginal basins during the  
336 first stage of the salinity crisis (Roveri *et al.* 2014). Therefore, the  $^{87}\text{Sr}/^{86}\text{Sr}$  values of the  
337 Nakhon Sawan gypsum samples (Table 3) are inferred to reflect those of coeval ocean  
338 water.

339

#### 340 AGES OF THE ANDESITE INTRUSION

341 Zircons from the andesitic dike sample 13112605 yield U–Pb ages that range from  
342  $231.7 \pm 5.8$  to  $436.7 \pm 11.8$  Ma, with a single older Proterozoic age of  $2404.7 \pm 63.9$  Ma  
343 ( $n = 33$ , Supporting Information Table S2). The results of U–Pb dating of zircons from  
344 the andesitic dike that are younger than 500 Ma are shown on a concordia diagram (Fig.  
345 S1 A) and on a relative age probability diagram (Fig. S1 B). The age distribution of  
346 zircons from the dike sample is characterized by multiple peaks, suggesting a variety of  
347 origins for the zircon grains (Fig. S1 B). The youngest age cluster is between 232 and  
348 245 Ma (mean = 238.3 Ma), and the age cluster with the most intense peak ranges from  
349 274 to 302 Ma (weighted mean age =  $291.3 \pm 4.8$  Ma). Zircons from the dikes could  
350 have U–Pb ages that are older than the intrusion age if inherited and xenocrystic zircons  
351 were entrained into the dike magma (e.g., Keay *et al.* 1999; Shinjoe *et al.* 2003; Wang *et al.*  
352 2012). The andesitic dike sample 13112605 contains xenoliths as well as xenocrysts  
353 of quartz. The quartz crystals seem to have originated from broken quartz veins and  
354 xenolithic materials of the host rocks. Assuming that the youngest cluster of zircon ages  
355 represents the timing of intrusion, the age of the andesitic dike is roughly 240 Ma, or

356 early Middle Triassic, and this corresponds to the timing of a major episode of  
357 volcanism in the Sukhothai Arc (Barr *et al.* 2000; Srichan *et al.* 2009; Hara *et al.* 2013).  
358 Trace element compositions of the igneous dike normalized to CI chondrite (Anders and  
359 Grevesse, 1989) are best fit to that of the andesite rocks within the Sukhothai Arc  
360 described by Srichan *et al.* (2009) (Fig. S2). These results indicate that the andesitic  
361 dikes that intruded the gypsum deposits were part of the volcanic activity within the  
362 Sukhothai Arc during the Early to Middle Triassic.

363

## 364 **DISCUSSION**

### 365 AGES OF THE SULFATE DEPOSITS

366 Both the  $\delta^{34}\text{S}$  and  $^{87}\text{Sr}/^{86}\text{S}$  values of our gypsum and anhydrite samples from the  
367 Nakhon Sawan area are likely to reflect the values of the coeval ocean water from  
368 which the sulfate evaporite was originally precipitated. This gives us a critical clue for  
369 constraining the age of the sulfate evaporite, because we can compare our data with the  
370 compiled database of the secular variation of isotopic compositions of strontium (e.g.,  
371 McArthur *et al.* 2012) and sulfur (e.g., Kampschulte & Strauss 2004) (Fig. 7).

372 The  $^{87}\text{Sr}/^{86}\text{Sr}$  values of our gypsum and anhydrite samples range from 0.70810 to  
373 0.70817 (Table 3). During the last 500 Ma, this range of  $^{87}\text{Sr}/^{86}\text{Sr}$  values in ocean water  
374 has been observed in eight time windows, at ca. 26, 300, 326, 353–355, 364–367, 403–  
375 406, 439, and 454 Ma (Fig. 7). Given the fact that both the gypsum and anhydrite  
376 deposits are cut by a Middle Triassic andesitic dikes (~240 Ma), the youngest time  
377 window of 26 Ma is excluded, and the likelihood is that the deposits are at least  
378 Carboniferous or older.

379 The  $\delta^{34}\text{S}$  values of our gypsum samples range between 15.86 and 16.26‰ (Table 2).

380 During the last 500 Ma, ocean water  $\delta^{34}\text{S}$  values have changed in a range between  
381 +10‰ and +30‰ (Fig. 7) (Kampschulte & Strauss 2004; Paytan & Gray 2012). The  
382 recorded  $\delta^{34}\text{S}$  values of evaporitic sulfates and carbonate-associated sulfates show  
383 relatively higher values (greater than +20‰) from the Cambrian through the Early  
384 Mississippian, and in the early Permian (Fig. 7). The  $\delta^{34}\text{S}$  values of our gypsum samples  
385 fit well with the ocean water sulfate  $\delta^{34}\text{S}$  curves between the Early Mississippian and  
386 Middle Pennsylvanian, and between the Late Permian and younger ages, but the latter  
387 possibility can be excluded by the presence of the cross-cutting Middle Triassic  
388 andesitic dikes and Sr isotope data.

389 There is a point on the ocean water curves that fits both the  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\delta^{34}\text{S}$   
390 values of our gypsum and anhydrite samples, and that point (~326 Ma) is in the  
391 Serpukhovian (331-323 Ma) of the late Mississippian/the early Carboniferous (Fig. 7).  
392 The Tournaisian (359–347 Ma) of the Early Mississippian and the Gzhelian (304-229  
393 Ma) of the Late Pennsylvanian are other possibilities. However, the ocean water sulfate  
394  $\delta^{34}\text{S}$  values for the Tournaisian are significantly higher than those of our gypsum  
395 samples, and although there are no available data for evaporitic sulfate  $\delta^{34}\text{S}$  values in  
396 the Late Pennsylvanian, the Gzhelian  $\delta^{34}\text{S}$  values for carbonate-associated sulfates are  
397 lower than our gypsum values (Fig. 7). Therefore, we suggest that the Tournaisian and  
398 the Gzhelian are less likely ages for our gypsum and anhydrite samples than the  
399 Serpukhovian.

400

#### 401 ENVIRONMENTAL IMPLICATIONS

402 Previously, Ueno & Charoentitirat (2011) concluded the age of the sulfate mineral  
403 deposits in the Nakhon Sawan area fell on the Mississippian–Pennsylvanian boundary,

404 based on fossils in carbonate rocks in the vicinity. Our isotopic results are consistent  
405 with that age assessment, and we suggest that the sulfate evaporites were precipitated  
406 from seawater at the end of the Mississippian (Serpukhovian), in a shallow basin on a  
407 carbonate platform (Khao Khwang Platform) at the margin of the Indochina Block. The  
408 global seawater  $^{87}\text{Sr}/^{86}\text{Sr}$  record shows a gradual increase through the Serpukhovian  
409 (Fig. 7). Our  $^{87}\text{Sr}/^{86}\text{Sr}$  data from the core GYP13 does not show any change with depth  
410 (Table 3). This suggests that the sulfate rock body has precipitated within short duration,  
411 probably much shorter than the residence time of Sr in the ocean (4-5 million years at  
412 present).

413 Our results indicate that this site on the Khao Khwang Platform was subject to arid  
414 climatic conditions during the Serpukhovian. According to Metcalfe (2006), the  
415 Indochina Block was located between the paleoequator and  $10^\circ\text{N}$  from the early  
416 Carboniferous to the early Permian. The emergence of arid climatic conditions in this  
417 equatorial region might be explained by the migration or meandering of the  
418 inter-tropical convergent zone (ITCZ) away from the equator during this time, or  
419 alternatively it could have been the result of a unique distribution of land and sea, and  
420 circulation of the atmosphere and ocean in this specific location, rather like present-day  
421 East Africa, where hyper-arid conditions prevail within an equatorial region (e.g., Le  
422 Hou  rou 2008).

423 On the western margin of the Indochina Block, similar sulfate deposits (gypsum–  
424 anhydrite) are also known in the Loei–Wang Saphung area of northeast Thailand  
425 (Surakotra *et al.* 2005). Those deposits have been assigned a late Moscovian (Middle  
426 Pennsylvanian) age, based on their stratigraphic relations with fossil-bearing limestones  
427 in borehole material (Fontaine *et al.* 1997; Surakotra *et al.* 2005), and this age is slightly

428 younger than that of the Nakhon Sawan sulfate deposits. In the Loei–Wang Saphung  
429 area, mixed siliciclastic–carbonate deposition prevailed throughout most of  
430 Pennsylvanian time, and normal open-marine conditions were common (Ueno &  
431 Charoentitirat 2011). Moreover, it is well known that the Early Pennsylvanian  
432 (Moscovian in particular) corresponds to a period of transgression in the record of  
433 long-term sea-level change (e.g., Haq & Schutter 2008). These environmental settings  
434 seem irreconcilable with the formation of a restricted hypersaline basin and the resultant  
435 sulfate evaporitic deposits on the shelf of the Indochina Block. As inferred by Ueno &  
436 Charoentitirat (2011), it seems more plausible to correlate the sulfate rock body in the  
437 Loei–Wang Saphung area with that at Nakhon Sawan, given our present state of  
438 knowledge. Further investigation is required to more precisely constrain the age of the  
439 Loei–Wang Saphung deposits, using the same methods as those described in this paper.

440

#### 441 **CONCLUSIONS**

442 The sulfate mineral deposits in the Nakhon Sawan area, central Thailand, are cut by  
443 andesitic dikes, and U–Pb dating of zircons from the dike rocks indicates emplacement  
444 in the Middle Triassic (~240 Ma). The dikes are probably related to regional magmatic  
445 activity in the Sukhothai Arc during the Early to Middle Triassic. The sulfate deposits  
446 are made up of thinly bedded anhydrite in the lower part, and heavily deformed massive  
447 and nodular gypsum rocks in the upper part, which is rehydrated from the anhydrite.  
448 The anhydrite was formed as a diagenetic product during burial through the dehydration  
449 of the original pristine gypsum that had been precipitated from seawater in a shallow  
450 lagoon or shelf environment. The sulfur and strontium isotopic compositions fit well  
451 with the seawater values for Serpukhovian time (~326 Ma).

452       Based on various lines of evidence, we interpret the history of the sulfate mineral  
453 deposits as follows: 1) precipitation of pristine gypsum from seawater on the floor of a  
454 shallow lagoon or shelf on the Khao Khwang Platform during the Serpukhovian (Late  
455 Mississippian); 2) transformation of the pristine gypsum into anhydrite through  
456 dehydration at an early stage of burial; 3) emplacement of andesitic dikes into the  
457 sulfate deposits during the Middle Triassic; and 4) rehydration of the top part of  
458 anhydrite to form secondary gypsum near the surface following exhumation.

459

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- 651

## 652 TABLES

653 **Table 1.** Major and trace element concentrations of the gypsum and anhydrite samples measured by ICP–MS.

Sample name	Location	Lithology	Mg ( $\mu\text{g g}^{-1}$ )	Al ( $\mu\text{g g}^{-1}$ )	K ( $\mu\text{g g}^{-1}$ )	Ca (wt.%)	Sc ( $\mu\text{g g}^{-1}$ )	Ti ( $\mu\text{g g}^{-1}$ )	V ( $\mu\text{g g}^{-1}$ )	Cr ( $\mu\text{g g}^{-1}$ )	Mn ( $\mu\text{g g}^{-1}$ )	Fe ( $\mu\text{g g}^{-1}$ )	Co ( $\mu\text{g g}^{-1}$ )	Ni ( $\mu\text{g g}^{-1}$ )	Cu ( $\mu\text{g g}^{-1}$ )	Zn ( $\mu\text{g g}^{-1}$ )	Sr ( $\mu\text{g g}^{-1}$ )	Y ( $\mu\text{g g}^{-1}$ )	Ba ( $\mu\text{g g}^{-1}$ )	Pb ( $\mu\text{g g}^{-1}$ )	U ( $\mu\text{g g}^{-1}$ )
JTG <sup>†</sup>	Saline di Trapani, Sicily	Gypsum	625	158	291	22.6	0.485	682	0.377	0.762	7.16	76.1	1.11	4.48	0.399	0.386	1511	0.0815	1.30	2.51	0.0264
13112601	Chao Phraya Mueang Rae	Gypsum	141	183	75.0	22.3	0.522	678	0.764	0.654	11.5	134	5.85	5.14	1.82	1.49	745	0.140	1.44	0.131	0.300
13112602	Chao Phraya Mueang Rae	Gypsum	36.3	52.6	54.3	21.9	0.385	664	0.549	0.173	2.55	68.0	1.04	4.40	0.143	<i>n.d.</i>	785	0.174	1.26	0.0267	0.0320
13112603	Chao Phraya Mueang Rae	Gypsum	27.3	174	40.8	25.0	0.331	673	0.264	0.159	0.953	66.1	0.861	4.98	0.172	0.118	491	0.0411	0.694	0.0736	0.00922
13112603'	Chao Phraya Mueang Rae	Gypsum	36.2	44.1	30.6	21.6	0.311	644	0.476	0.256	3.44	57.9	0.611	3.92	0.065	<i>n.d.</i>	560	0.0780	0.911	0.0222	0.0893
13112604	K-Mining	Gypsum	29.0	106	19.3	19.5	0.232	529	0.276	0.082	1.22	53.0	0.374	3.54	<i>n.d.</i>	<i>n.d.</i>	810	0.0498	0.913	0.0193	0.0383
GYP13 12-13	SCG Mine (core)	Gypsum	<i>n.d.</i>	251	70.9	22.5	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	2.20	444	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	247	<i>n.a.</i>	2.173	<i>n.a.</i>	<i>n.a.</i>
GYP13 15-16	SCG Mine (core)	Gypsum	1932	20.0	210	22.4	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	5.30	353	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	538	<i>n.a.</i>	0.572	<i>n.a.</i>	<i>n.a.</i>
GYP13 22-23	SCG Mine (core)	Gypsum	<i>n.d.</i>	<i>n.d.</i>	156	23.3	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.d.</i>	331	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	696	<i>n.a.</i>	0.418	<i>n.a.</i>	<i>n.a.</i>
GYP13 31-32	SCG Mine (core)	Gypsum	38.5	<i>n.d.</i>	233	24.0	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.d.</i>	359	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	606	<i>n.a.</i>	0.158	<i>n.a.</i>	<i>n.a.</i>
GYP13 37-38	SCG Mine (core)	Anhydrite	<i>n.d.</i>	<i>n.d.</i>	286	25.4	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	2.32	347	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	742	<i>n.a.</i>	0.673	<i>n.a.</i>	<i>n.a.</i>
GYP13 43-44	SCG Mine (core)	Anhydrite	<i>n.d.</i>	<i>n.d.</i>	150	25.7	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	11.6	361	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	794	<i>n.a.</i>	0.138	<i>n.a.</i>	<i>n.a.</i>
GYP13 49-50	SCG Mine (core)	Anhydrite	87.7	<i>n.d.</i>	196	28.0	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	6.15	407	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	1000	<i>n.a.</i>	0.143	<i>n.a.</i>	<i>n.a.</i>
GYP13 57-58	SCG Mine (core)	Anhydrite	6.64	<i>n.d.</i>	182	29.5	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	0.806	418	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	<i>n.a.</i>	1063	<i>n.a.</i>	0.355	<i>n.a.</i>	<i>n.a.</i>

654

655 <sup>†</sup> JAMSTEC in-house standard of gypsum rock reference material taken from the Saline di Trapani, Sicily.656 *n.d.*: not detected, *n.a.*: not analyzed. Analytical uncertainties (as relative standard deviation) for the bulk rock analysis were better than

657 3% for major elements, and better than 5% for trace elements.

658

659 **Table 2.** Sulfur concentrations and stable sulfur isotopic compositions of the gypsum  
660 samples from the quarries in the Chon Daen area, east of Nakhon Sawan.

Sample name	Location	Lithology	Total S (wt.%)	S.D.	$\delta^{34}\text{S}$ (‰ V-CDT)	S.D.
13112601	Chao Phraya Mueang Rae	Gypsum	16.3	1.3	15.855	0.199
13112602	Chao Phraya Mueang Rae	Gypsum	16.5	1.5	16.171	0.100
13112603	Chao Phraya Mueang Rae	Gypsum	17.1	1.4	16.151	0.091
13112604	K-Mining	Gypsum	16.6	2.3	16.258	0.123

661

662

663 **Table 3.** Sr isotopic ratios of the gypsum and anhydrite samples from the quarries in the  
 664 Chon Daen area, east of Nakhon Sawan.

Sample name	Location	Lithology	$^{87}\text{Sr}/^{86}\text{Sr}$	2 SE
JTG <sup>†</sup>	Saline di Trapani, Sicily	Gypsum	0.709167	0.000011
13112602	Chao Phraya Mueang Rae	Gypsum	0.708110	0.000010
13112603	Chao Phraya Mueang Rae	Gypsum	0.708157	0.000011
13112603'	Chao Phraya Mueang Rae	Gypsum	0.708150	0.000008
13112604	K-Mining	Gypsum	0.708171	0.000010
GYP13 12-13	SCG Mine (core)	Gypsum	0.708156	0.000008
GYP13 15-16	SCG Mine (core)	Gypsum	0.708156	0.000007
GYP13 22-23	SCG Mine (core)	Gypsum	0.708143	0.000014
GYP13 31-32	SCG Mine (core)	Gypsum	0.708169	0.000009
GYP13 37-38	SCG Mine (core)	Anhydrite	0.708152	0.000009
GYP13 43-44	SCG Mine (core)	Anhydrite	0.708144	0.000008
GYP13 49-50	SCG Mine (core)	Anhydrite	0.708136	0.000008
GYP13 57-58	SCG Mine (core)	Anhydrite	0.708144	0.000009

665

666 <sup>†</sup> JAMSTEC in-house standard of gypsum rock reference material taken from the Saline  
 667 di Trapani, Sicily.

668 **FIGURE CAPTIONS**

669 **Fig. 1.** Google Earth images of the study site. (A) Regional map showing the location of  
670 the study area. (B) Air photo of northeastern Nakhon Sawan Province showing the  
671 locations of the three quarries. (C) Air photo of the K-Mining quarry showing sampling  
672 locations. (D) Air photo of the Chao Phraya Mueang Rae quarry showing sampling  
673 locations. (E) Air photo of the SCG Mine quarry showing GYP13 drilling site.

674

675 **Fig. 2.** (A) Geotectonic subdivision of Thailand (Ueno & Charoentitirat 2011; Ueno *et*  
676 *al.* 2012). (B) Distribution of Carboniferous and Permian sedimentary rocks in central  
677 Thailand (DMR 1999; Ueno & Charoentitirat 2011). The location of the study site is  
678 indicated.

679

680 **Fig. 3.** Photographs of outcrops. (A) Nodular microcrystalline gypsum rock in the Chao  
681 Phraya Mueang Rae quarry (location is given in Fig. 1 D) showing sample locations for  
682 13112601, 13112602, 13112603 and 13112603', and showing an andesitic dike  
683 (horizontal band in the middle of the image) and strong deformation of vertical nodules  
684 (lower part of the image). (B) Andesitic dike (13112605) that intruded massive  
685 microcrystalline gypsum rock in the K-Mining quarry (location is given in Fig. 1 C).

686 Scale bars are 1 m.

687

688 **Fig. 4.** (A) Lithologic column of well GYP13 at SCG Mine. Drill site is given in Fig. 1  
689 E. (B) Split surfaces of the drilled core samples of GYP13. Sampling position of each  
690 piece is shown in (A).

691

692 **Fig. 5.** Plane polarized photomicrographs of gypsum and andesitic intrusions. (A)  
693 Gypsum crystals and a folded and sheared silicate lamina in a slightly thick section of  
694 sample 13112601. (B) A contact between an andesitic dike and microcrystalline gypsum  
695 in sample 13112602. (C) Gypsum crystals in a slightly thick section of sample  
696 13112603. (D) Andesitic dike in sample 13112605. Gy, gypsum; And, andesitic  
697 material; Cal, calcite vein; Chl, chlorite; Dac, dacitic material; Hbl, hornblende  
698 (chloritized); Pl, plagioclase (saussuritized); Qz, quartz.  
699

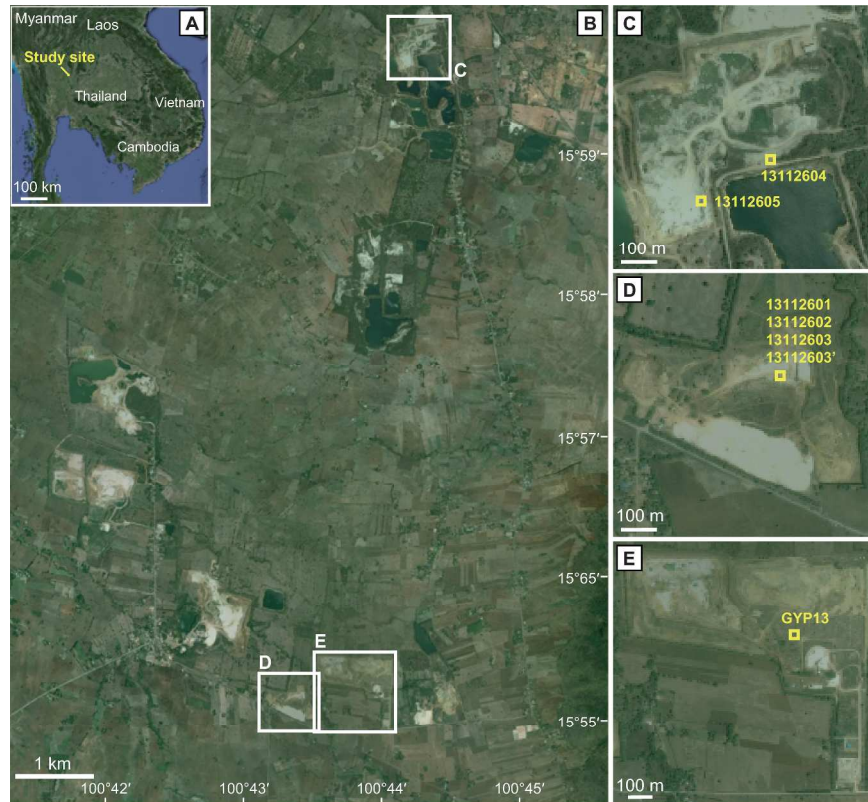
700 **Fig. 6.** Element maps showing the contact between sulfate minerals and an andesitic  
701 dike (sample 13112602). The distributions of elements are shown as intensities of  
702 characteristic X-rays for each element. Each scale bar is 3 mm.  
703

704 **Fig. 7.** Secular variations of strontium ( $^{87}\text{Sr}/^{86}\text{Sr}$ , middle panel) and sulfate sulfur ( $\delta^{34}\text{S}$ ,  
705 bottom panel) isotopic compositions in ocean water from 400 to 200 Ma. The  $^{87}\text{Sr}/^{86}\text{Sr}$   
706 values (green curve) are from LOWESS (data compiled by McArthur *et al.* 2012), and  
707 the  $\delta^{34}\text{S}$  values of carbonate-associated sulfates (blue curve) and sulfate evaporites (red)  
708 were compiled by Kampschulte & Strauss (2004). Horizontal lines indicate the data  
709 range for the Nakhon Sawan gypsum samples. The inset in each panel shows secular  
710 variations in isotopic compositions during the last 500 Ma (data sources are the same as  
711 given above).  
712

713 **SUPPORTING INFORMATION**

714 Additional Supporting Information may be found in the online version of this article at  
715 the publisher's web-site:

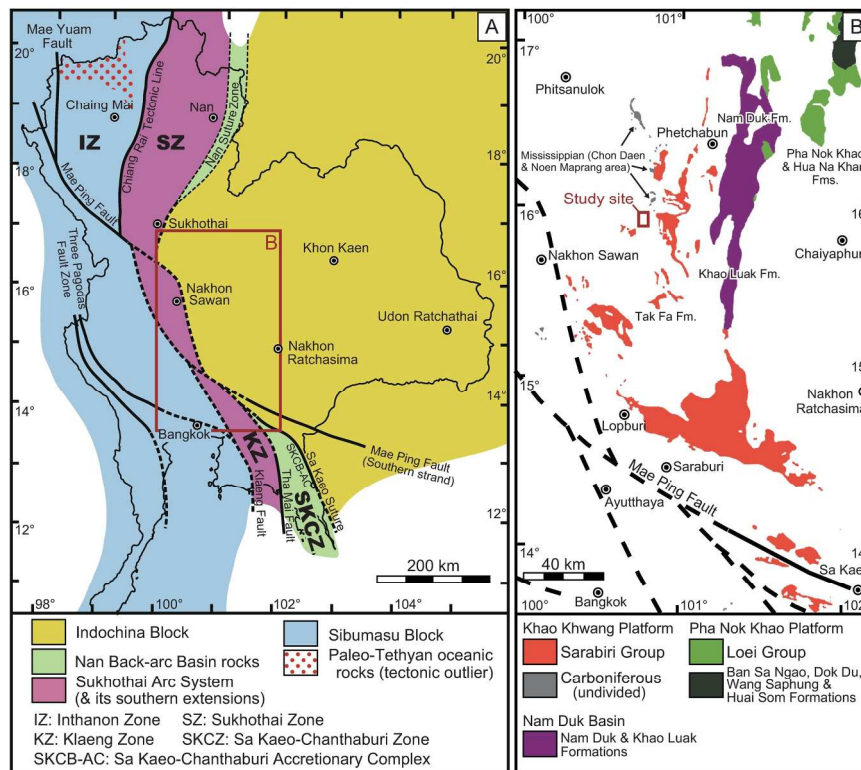
716



Kuroda et al. Fig. 1

Fig. 1. Google Earth images of the study site. (A) Regional map showing the location of the study area. (B) Air photo of northeastern Nakhon Sawan Province showing the locations of the three quarries. (C) Air photo of the K-Mining quarry showing sampling locations. (D) Air photo of the Chao Phraya Mueang Rae quarry showing sampling locations. (E) Air photo of the SCG Mine quarry showing GYP13 drilling site.

Fig. 1  
284x393mm (300 x 300 DPI)



Kuroda et al. Fig. 2

Fig. 2. (A) Geotectonic subdivision of Thailand (Ueno & Charoentitirat 2011; Ueno et al. 2012). (B) Distribution of Carboniferous and Permian sedimentary rocks in central Thailand (DMR 1999; Ueno & Charoentitirat 2011). The location of the study site is indicated.

Fig. 2

201x154mm (300 x 300 DPI)

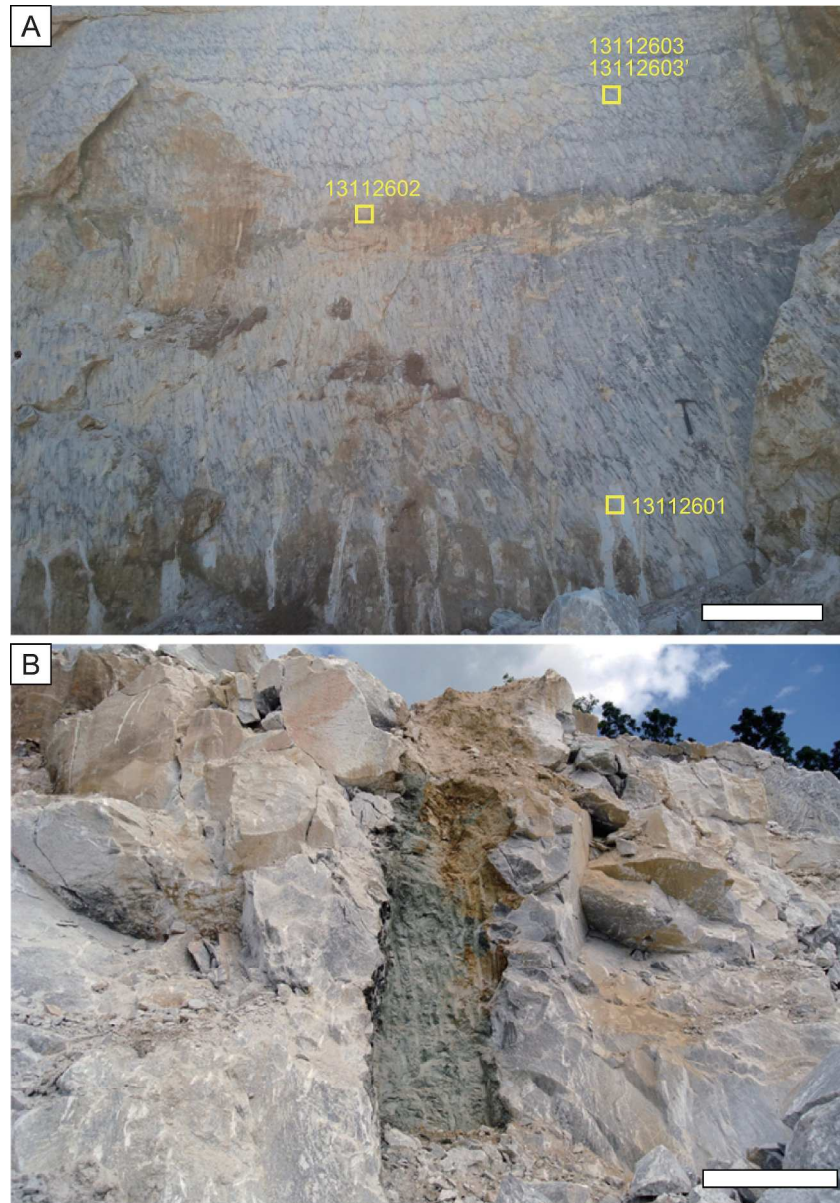
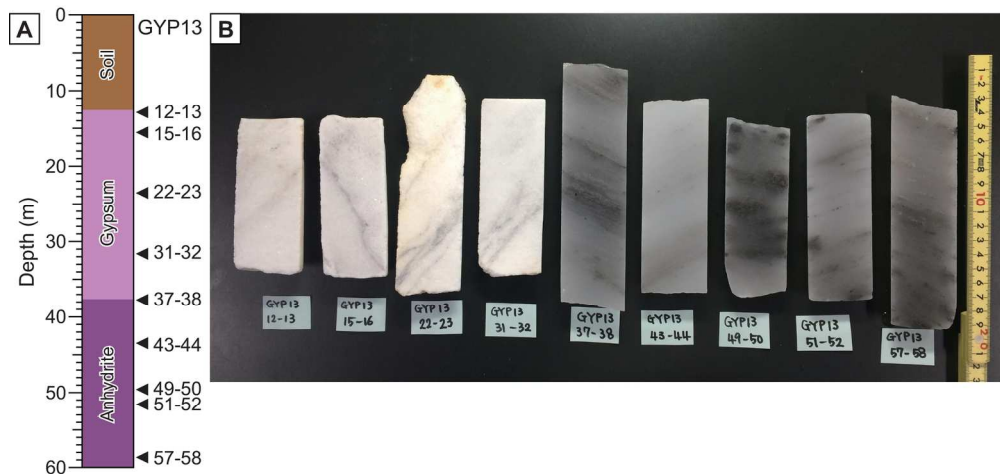


Fig. 3. Photographs of outcrops. (A) Nodular microcrystalline gypsum rock in the Chao Phraya Mueang Rae quarry (location is given in Fig. 1 D) showing sample locations for 13112601, 13112602, 13112603 and 13112603', and showing an andesitic dike (horizontal band in the middle of the image) and strong deformation of vertical nodules (lower part of the image). (B) Andesitic dike (13112605) that intruded massive microcrystalline gypsum rock in the K-Mining quarry (location is given in Fig. 1 C). Scale bars are 1 m.

Fig. 3  
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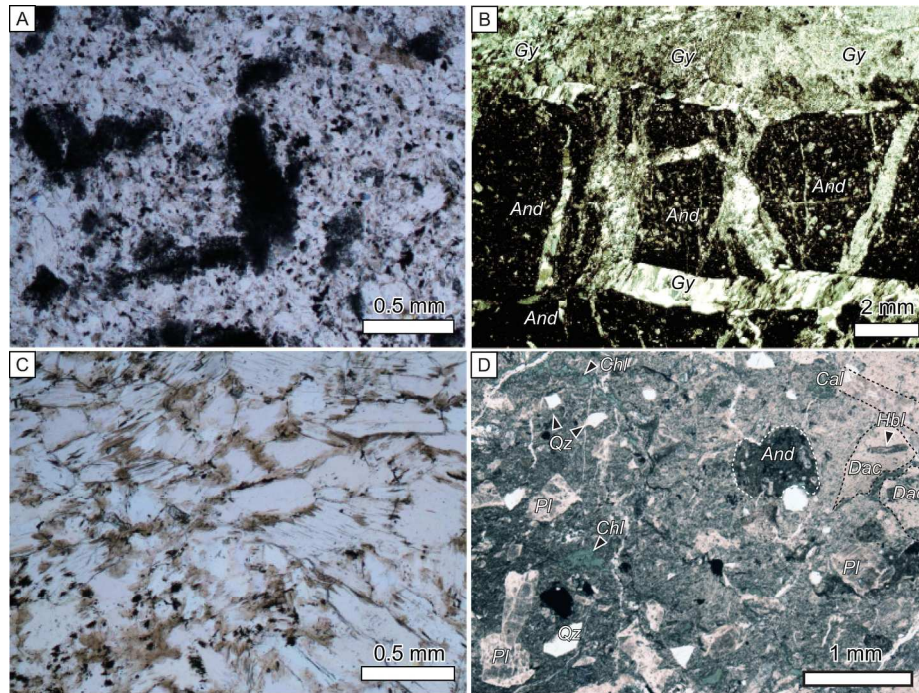


Kuroda et al. Fig. 4

Fig. 4. (A) Lithologic column of well GYP13 at SCG Mine. Drill site is given in Fig. 1 E. (B) Split surfaces of the drilled core samples of GYP13. Sampling position of each piece is shown in (A).

Fig. 4

167x100mm (300 x 300 DPI)

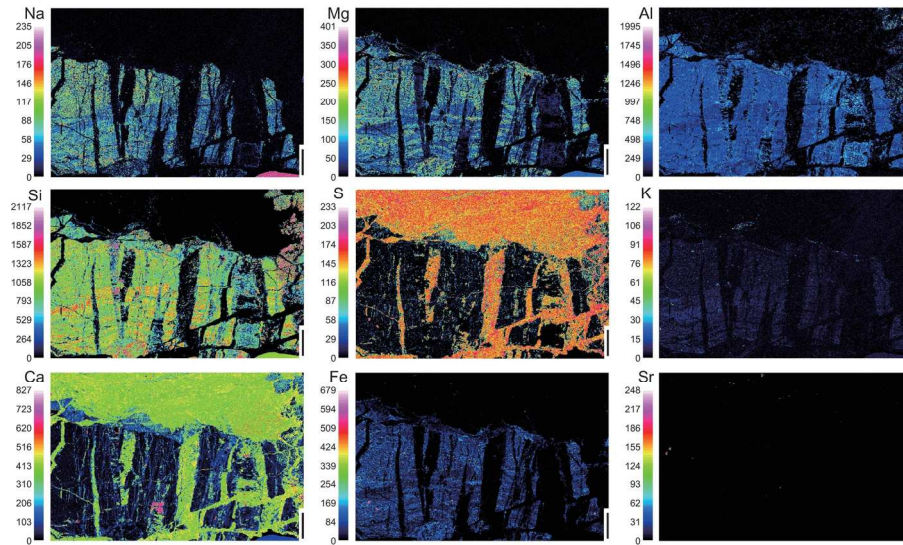


Kuroda et al. Fig. 5

Fig. 5. Plane polarized photomicrographs of gypsum and andesitic intrusions. (A) Gypsum crystals and a folded and sheared silicate lamina in a slightly thick section of sample 13112601. (B) A contact between an andesitic dike and microcrystalline gypsum in sample 13112602. (C) Gypsum crystals in a slightly thick section of sample 13112603. (D) Andesitic dike in sample 13112605. Gy, gypsum; And, andesitic material; Cal, calcite vein; Chl, chlorite; Dac, dacitic material; Hbl, hornblende (chloritized); Pl, plagioclase (saussuritized); Qz, quartz.

Fig. 5

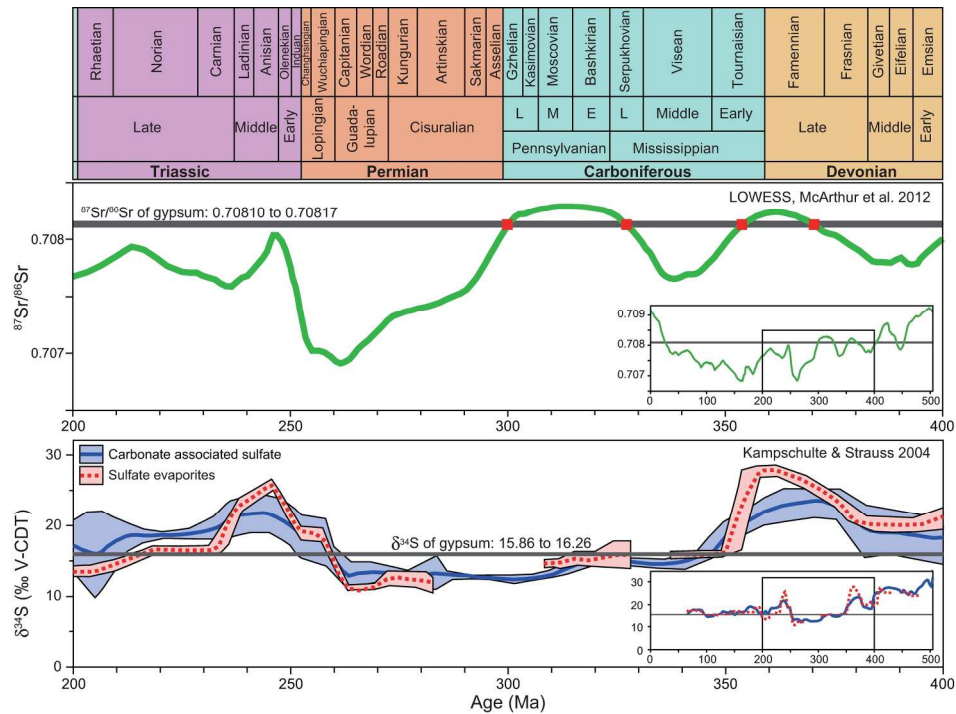
196x135mm (300 x 300 DPI)



Kuroda et al. Fig. 6

Fig. 6. Element maps showing the contact between sulfate minerals and an andesitic dike (sample 13112602). The distributions of elements are shown as intensities of characteristic X-rays for each element. Each scale bar is 3 mm.

Fig. 6  
173x114mm (300 x 300 DPI)



Kuroda et al. Fig. 7

Fig. 7. Secular variations of strontium ( $^{87}\text{Sr}/^{86}\text{Sr}$ , middle panel) and sulfate sulfur ( $\delta^{34}\text{S}$ , bottom panel) isotopic compositions in ocean water from 400 to 200 Ma. The  $^{87}\text{Sr}/^{86}\text{Sr}$  values (green curve) are from LOWESS (data compiled by McArthur et al. 2012), and the  $\delta^{34}\text{S}$  values of carbonate-associated sulfates (blue curve) and sulfate evaporites (red) were compiled by Kampschulte & Strauss (2004). Horizontal lines indicate the data range for the Nakhon Sawan gypsum samples. The inset in each panel shows secular variations in isotopic compositions during the last 500 Ma (data sources are the same as given above).

Fig. 7

205x145mm (300 x 300 DPI)

## SUPPORTING INFORMATION

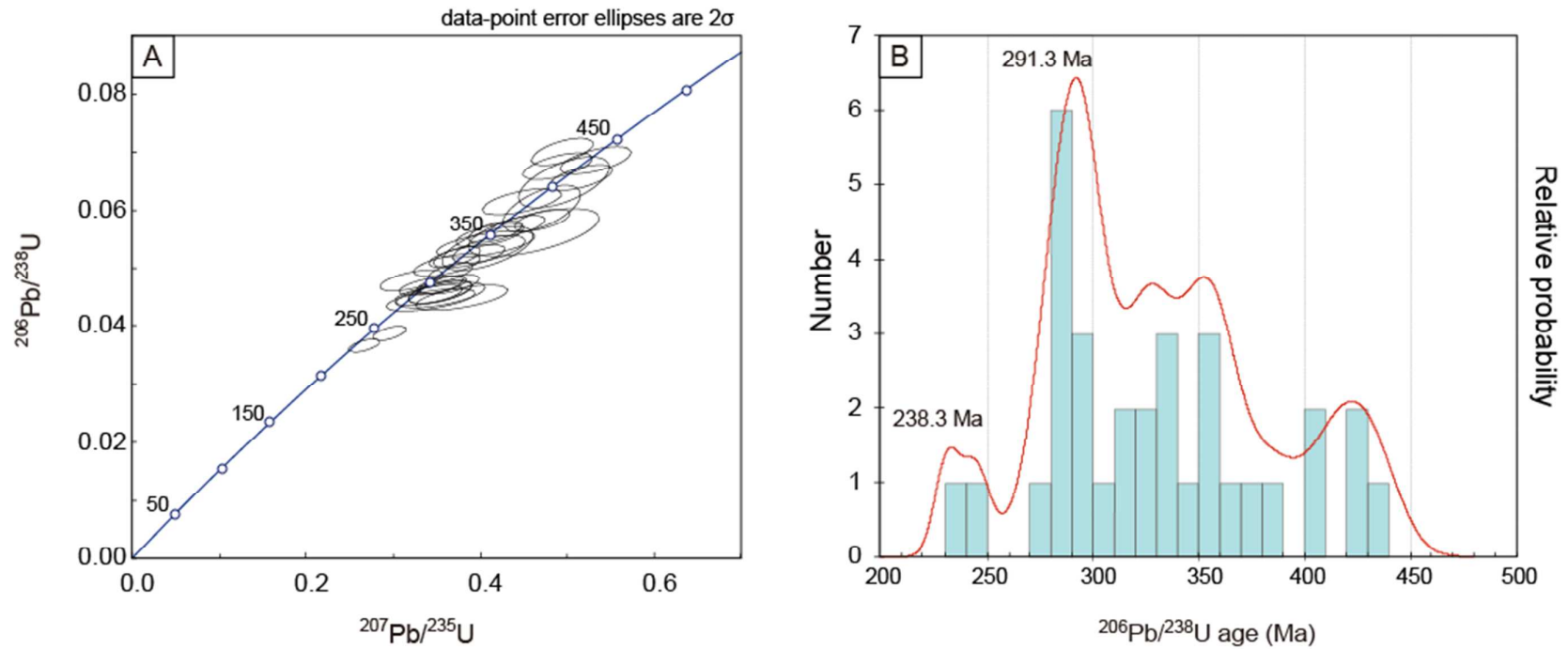
**Table S1.** Major and trace element compositions of the andesitic dike. Geochemical analyses of the dike were performed by X-ray fluorescence (XRF; Rigaku RIX3000) analysis of fused glass beads at Niigata University.

Major element	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub> *	MnO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	Total	L.O.I.
(wt. %)	59.91	0.49	15.26	5.39	0.12	2.33	7.02	1.37	0.51	0.07	100.29	7.2
Trace element	Cr	Nb	Ni	Rb	Sr	V	Y	Zr	Pb	Th		
( $\mu\text{g g}^{-1}$ )	107.2	12.3	3.89	3.2	11.65	537.95	102.7	35.305	105.42	13.056	4.399	

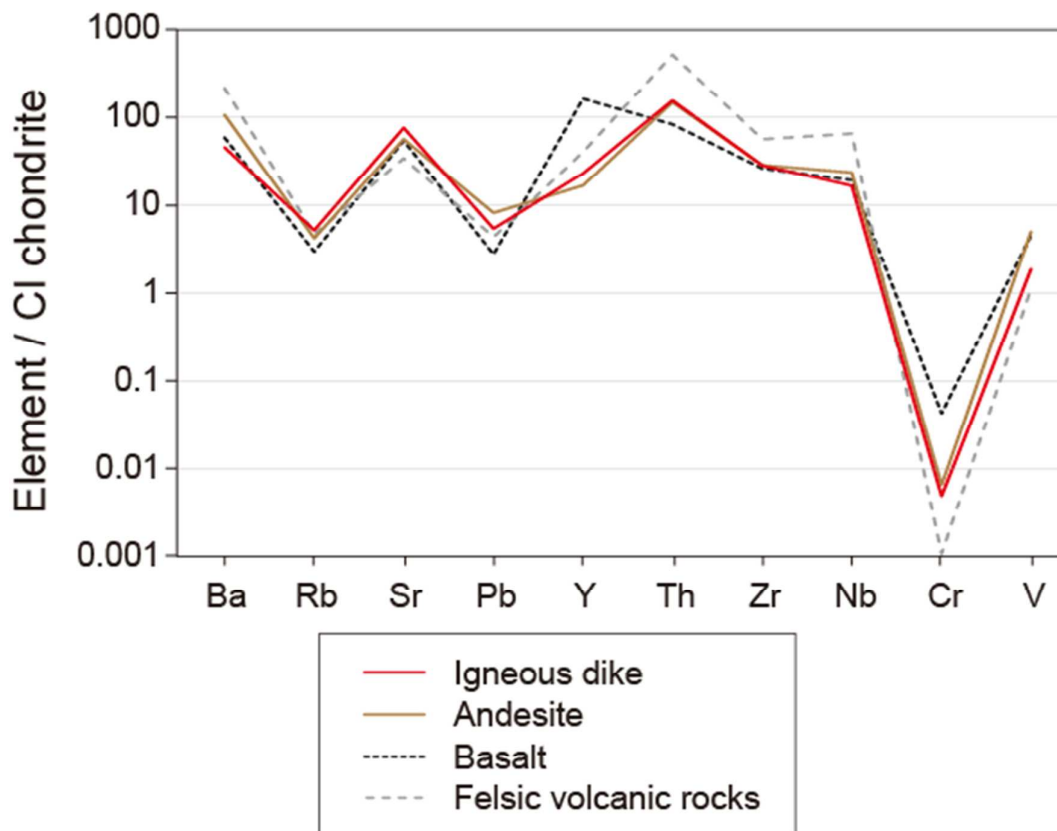
L.O.I.: Weight loss on ignition, Fe<sub>2</sub>O<sub>3</sub>\* is total iron as Fe<sub>2</sub>O<sub>3</sub>.

**Table S2.** U–Pb isotopic data (concordia data) for zircon crystals from the andesitic dike (sample 112605). Analyses were performed using LA–ICP–MS.

Grain Number	Th/U	$^{206}\text{Pb}/^{238}\text{U}$	Error 2 $\sigma$	$^{207}\text{Pb}/^{235}\text{U}$	Error 2 $\sigma$	$^{238}\text{U}$ – $^{206}\text{Pb}$ age (Ma)	Error 2 $\sigma$	$^{235}\text{U}$ – $^{207}\text{Pb}$ age (Ma)	Error 2 $\sigma$
01	0.70	0.0520 ± 0.0016		0.385 ± 0.036		327.1 ± 10.2		330.6 ± 30.6	
02	0.91	0.0471 ± 0.0013		0.354 ± 0.022		296.7 ± 8.1		307.8 ± 19.2	
04	0.90	0.0583 ± 0.0016		0.471 ± 0.026		365.1 ± 9.8		392.1 ± 21.6	
05	0.87	0.0676 ± 0.0019		0.489 ± 0.032		421.5 ± 11.7		404.3 ± 26.8	
06	0.55	0.0520 ± 0.0014		0.373 ± 0.022		326.7 ± 8.8		322.0 ± 19.0	
07	1.03	0.0686 ± 0.0019		0.533 ± 0.033		427.7 ± 11.9		433.6 ± 27.2	
08	0.48	0.0701 ± 0.0019		0.494 ± 0.029		436.7 ± 11.8		407.9 ± 23.7	
10	0.79	0.0541 ± 0.0018		0.406 ± 0.046		339.6 ± 11.6		346.3 ± 39.4	
11	0.40	0.0656 ± 0.0018		0.516 ± 0.029		409.8 ± 11.3		422.4 ± 24.0	
12	0.44	0.4521 ± 0.0120		9.846 ± 0.382		2,404.7 ± 63.9		2,420.3 ± 93.9	
13	0.89	0.0505 ± 0.0015		0.362 ± 0.031		317.8 ± 9.7		313.6 ± 27.0	
14	0.61	0.0445 ± 0.0016		0.343 ± 0.042		280.7 ± 9.9		299.4 ± 36.4	
16	0.68	0.0453 ± 0.0014		0.340 ± 0.032		285.7 ± 9.0		297.5 ± 28.1	
17	0.30	0.0562 ± 0.0015		0.417 ± 0.025		352.6 ± 9.7		354.2 ± 21.5	
18	0.66	0.0557 ± 0.0016		0.411 ± 0.032		349.2 ± 10.3		349.7 ± 27.0	
19	0.79	0.0493 ± 0.0013		0.366 ± 0.021		310.4 ± 8.4		316.3 ± 18.1	
20	0.57	0.0479 ± 0.0015		0.321 ± 0.030		301.6 ± 9.4		282.9 ± 26.3	
21	0.61	0.0615 ± 0.0018		0.448 ± 0.037		384.5 ± 11.6		375.8 ± 31.1	
22	1.18	0.0387 ± 0.0010		0.296 ± 0.016		244.9 ± 6.5		263.2 ± 14.0	
23	0.77	0.0472 ± 0.0014		0.364 ± 0.028		297.2 ± 8.8		315.4 ± 24.5	
25	0.66	0.0470 ± 0.0014		0.346 ± 0.032		296.3 ± 8.8		301.7 ± 27.6	
26	0.89	0.0366 ± 0.0009		0.267 ± 0.014		231.7 ± 5.8		239.9 ± 13.0	
32	0.35	0.0572 ± 0.0015		0.443 ± 0.026		358.9 ± 9.1		372.3 ± 21.8	
34	0.68	0.0449 ± 0.0018		0.379 ± 0.043		283.1 ± 11.2		326.2 ± 37.2	
35	0.72	0.0458 ± 0.0016		0.354 ± 0.030		288.7 ± 10.2		307.9 ± 26.0	
36	0.67	0.0459 ± 0.0016		0.354 ± 0.030		289.1 ± 10.2		307.4 ± 25.9	
38	0.67	0.0453 ± 0.0018		0.355 ± 0.042		285.8 ± 11.3		308.1 ± 36.3	
40	1.01	0.0533 ± 0.0019		0.410 ± 0.034		334.5 ± 11.8		349.0 ± 29.1	
41	0.82	0.0534 ± 0.0030		0.409 ± 0.043		335.3 ± 19.1		347.8 ± 36.3	
42	0.38	0.0648 ± 0.0036		0.496 ± 0.043		404.8 ± 22.5		409.1 ± 35.1	
43	0.46	0.0602 ± 0.0033		0.471 ± 0.036		377.0 ± 20.7		391.6 ± 30.3	
46	0.11	0.0561 ± 0.0033		0.461 ± 0.061		352.0 ± 21.0		385.3 ± 51.3	
47	0.47	0.0434 ± 0.0028		0.378 ± 0.069		274.1 ± 17.7		325.7 ± 59.5	



**Fig. S1.** Analytical data for zircons younger than 500 Ma from the andesitic dike sample 13112605. (A) Concordia diagram. (B) Relative probability plot and age distribution histogram. Analytical errors are present at 2 $\sigma$ .



**Fig. S2.** Trace element composition of the andesitic dike normalized against CI chondrite (Anders and Grevesse 1989). Geochemical data for volcanic rocks in the Sukhothai Arc are from Srichan *et al.* (2009).

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